Environmentally dependent dust chemistry of a super Asian dust storm in
March 2010: observation and simulation

Qiongzhen Wang1, 2, Xinyi Dong3, Joshua S. Fu3, Jian Xu1, Congrui Deng1, Yilun
Jiang1, Qingyan Fu4, Yanfen Lin4, Kan Huang1,5*, Guoshun Zhuang1*
1Center for Atmospheric Chemistry Study, Shanghai Key Laboratory of Atmospheric
Particle Pollution and Prevention (LAP3), Department of Environmental Science and
Engineering, Fudan University, Shanghai, 200433, P. R. China
2Environmental Science Research & Design Institute of Zhejiang Province, Hangzhou,
310007, P. R. China
3Department of Civil and Environmental Engineering, University of Tennessee,
Knoxville, TN, 37996, USA
4Shanghai Environmental Monitoring Center, Shanghai, 200030, China
5Institute of Atmospheric Sciences, Fudan University, Shanghai, 200433, P. R. China

Correspondence: huangkan@fudan.edu.cn; gzhuang@fudan.edu.cn

Abstract
Near surface and vertical in situ measurements of atmospheric aerosols were
conducted in Shanghai during March 19-27, 2010 to explore the transport and
chemical evolution of dust aerosols in a super dust storm. An air quality model with
optimized physical dust emission scheme and newly implemented dust chemistry was
utilized to study the impact of dust chemistry on regional air quality. Two
discontinuous dust periods were observed with one travelling over Northern China
(DS1) and the other passing over the coastal regions of Eastern China (DS2). Stronger
mixing extents between dust and anthropogenic emissions were found in DS2,
reflecting by the higher SO\textsubscript{2}/PM\textsubscript{10} and NO\textsubscript{2}/PM\textsubscript{10} ratios as well as typical pollution
elemental species such as As, Cd, Pb, and Zn. As a result, the concentrations of SO\textsubscript{4}^2-
and NO$_3^-$ and the ratio of Ca$^{2+}$/Ca were more elevated in DS2 than in DS1 but opposite for the [NH$_4^+$]/[SO$_4^{2-}$+NO$_3^-$] ratio, suggesting the heterogeneous reactions between calcites and acid gases were significantly promoted in DS2 due to the higher level of relative humidity and gaseous pollution precursors. Lidar observation showed a columnar effect on the vertical structure of aerosol optical properties in DS1 that dust dominantly accounted for ~80-90% of the total aerosol extinction from near the ground to ~700m. In contrast, the dust plumes in DS2 were refrained within lower altitudes while the extinction from spherics particles exhibited maximum at a high altitude of ~800m. The model simulation reproduced relatively consistent results with observations that strong impacts of dust heterogeneous reactions on secondary aerosol formation occurred in areas where the anthropogenic emissions were intensive. Compared to the sulfate simulation, the nitrate formation on dust is suggested to be improved in the future modeling efforts.

1. Introduction

Asian dust originating from the arid and semiarid areas in Mongolia and China can be transported for long distances, reaching Beijing (Sun et al., 2010), Shanghai (Fu et al., 2010), Xiamen (Zhao et al., 2011), Taiwan (Tsai et al., 2012; Tsai et al., 2014), and even as far as North America (Uno et al., 2009; Wu et al., 2015), exerting significant impacts on the air quality of both densely populated habitations and remote regions. Dust aerosols can significantly influence the regional/global climate directly by absorbing and scattering solar radiation (Bi et al., 2016) and also indirectly by influencing the formation of ice nuclei, cloud, and precipitation (Creamean et al., 2013; Li and Min, 2010; Wang et al., 2010). In addition, deposition of transported dust aerosols into the ocean can enhance phytoplankton blooms due to the existence of bioavailable iron (Wang et al., 2012; Zhuang et al., 1992), which indirectly impacts on global climate change.

The effects of dust aerosols on climate change depend critically on their physical
and chemical properties. Natural dust aerosols with limited contamination have low light-absorption, with single-scattering albedo of 0.91-0.97 at 500nm and 550nm (Bi et al., 2014; Uchiyama et al., 2005). During the long-range transport, dust aerosols are often modified by their mixing with anthropogenic emissions over the downwind areas (Fischer et al., 2011; Formenti et al., 2011; Huang et al., 2010b; Tobo et al., 2010), resulting in high uncertainties in evaluating the climatic effects of dust aerosols.

It was estimated that mineral dust had a radiative forcing of $-0.1 \pm 0.2 \, \text{Wm}^{-2}$ (IPCC, 2013), of which the uncertainty was as high as 200%. Obviously, the characteristics of dust aerosols and their evolution during the transport are not well understood.

In March 2010, a super dust storm swept China, invading extensive areas from Northern China to Southern China including Fujian and Guangdong provinces, and lasting for ~4 days from March 19 to 23 (Li et al., 2011). The dust plumes further extended to the South China Sea (Wang et al., 2011), Taiwan (Tsai et al., 2013), Korea (Tatarov et al., 2012), Japan (Zaizen et al., 2014), and even to North America (Wu et al., 2015). This dust storm was as strong as the one in March 20-21, 2002 and attracted considerable attentions. Lidar observations revealed that this super dust storm was transported within a low altitude (Tatarov et al., 2012; Wang et al., 2011), which could benefit the mixing and interaction between dust aerosols and anthropogenic pollutants. Indeed, modifications of dust aerosols during the transport of this dust storm were suggested based on in situ measurements. Zhao et al. (2011) displayed substantial increases of sulfate and nitrate in aerosols when the dust plumes arrived at Xiamen city of Fujian province, implying the mixing and interaction between dust aerosols and anthropogenic pollutants. Wang et al. (2011) indicated that the dust aerosols detected at the Dongshan Island over the South China Sea were mixed with anthropogenic and marine aerosols. Observations of this dust storm at Tsukuba and Mt. Haruna, Japan showed that most of the transported dust aerosols in lower altitudes were internally mixed with sulfate or seasalt (Zaizen et al., 2014).

Most of the studies on this super dust storm focused on investigating the dust
aerosols reaching Southeastern China and the South China Sea, and relied on single method, e.g. aerosol chemistry measurement, optical property inversion, or model simulation. In this study, we investigated this super dust storm in Shanghai, a coastal city in Eastern China. A synergy of measurement techniques was applied, including in situ measurements of pollutant gaseous precursors, aerosol with its major chemical components, and Lidar observation of aerosol optical properties. To corroborate the observational evidence, a regional numerical model was used to simulate the impact of dust chemistry on the perturbation of regional air quality. What interested us is that there were two discontinuous dust periods observed in Shanghai with distinctly different transport pathways, providing a great opportunity to study the chemical evolution of transported dust particles under different environmental conditions.

2. Methodology

2.1. Field measurement

2.1.1. Lidar observation

A dual-wavelength depolarization Lidar (Model:L2S-SMII) developed by the National Institute for Environmental Studies (NIES) of Japan was installed on the roof (~20m above ground level) of a teaching building on the campus of Fudan University in the Yangpu District of Shanghai (Fig. 1b). The Lidar measurement was performed every 15 min (at 00, 15, 30, and 45 minutes every hour) with a height resolution of 6 m. Attenuated backscattering coefficient (BSC) and depolarization ratio (DR) of particles at the wavelength of 532 nm were obtained by the measurement. Aerosol extinction coefficients were derived by the Fernald inversion method (Fernald, 1984) with the lidar ratio (extinction-to-backscatter ratio) set as 50 sr (Liu et al., 2002) in the inversion process. The total aerosol extinction coefficient can be split to non-spherical particle (dust particle) and spherical particle (mostly pollution particle) fractions based on the DR of particles. The splitting method was described in detail by Sugimoto et al. (2002) and Shimizu et al. (2004). More details about the Lidar system...
have been described in Huang et al. (2012).

2.1.2. Online particle and gases monitoring
Continuous PM$_{10}$ concentrations were measured by a TEOM (Tapered Element Oscillating Microbalance) 1405D monitor (Thermo Scientific, USA). Trace gases SO$_2$ and NO$_2$ were measured by a 43i SO$_2$ analyzer (Thermo Scientific, USA) and a 42i NO-NO$_2$-NOx analyzer (Thermo Scientific, USA), respectively. All the measured PM$_{10}$ and gases concentrations were averaged and used at intervals of 1hr in this study.

2.1.3. Aerosol sampling
TSP (Total Suspended Particles) samples were collected during March 19-27, 2010 at the Fudan observational site co-located with all the other instruments. The aerosol samples were collected for 12 hours (normally from 8:00 to 20:00LST (Local Standard Time) in daytime and from 20:00 to 8:00 LST of the next day in nighttime) or 24 hours (normally from 8:00 to 8:00 LST of the next day) on Whatman 41 filters (Whatman Inc., Maidstone, UK) by a medium-volume sampler (Beijing Geological Instrument-Dickel Co., Ltd.; model: TSP/PM$_{10}$/PM$_{2.5}$-2; flow rate: 77.59 L min$^{-1}$). All the samples were put in polyethylene plastic bags immediately after sampling and then reserved in a refrigerator. The filters were weighed before and after sampling using an analytical balance (Model: Sartorius 2004MP; reading precision: $10\mu$g) after stabilizing in constant temperature (20±1°C) and humidity (40±2%) for 48 hours. All the procedures were strictly quality controlled to avoid the possible contamination of the samples.

2.2. Chemical analysis
2.2.1. Ion analysis
One fourth of each aerosol sample and blank filters were extracted ultrasonically by
10 ml deionized water (18 MΩ cm\(^{-1}\)). Inorganic ions of \(\text{SO}_4^{2-}\), \(\text{NO}_3^-\), \(\text{Cl}^-\), \(\text{Na}^+\), \(\text{NH}_4^+\), \(\text{K}^+\), \(\text{Mg}^{2+}\), and \(\text{Ca}^{2+}\) were analyzed by an Ion Chromatography (Dionex ICS 3000, USA) with a separation column of Dionex Ionpac AS 11, a guard column of Dionex Ionpac AG 11, a self-regenerating suppressed conductivity detector of Dionex Ionpac ED50, and a gradient pump of Dionex Ionpac GP50. The detailed analytical procedures can be found in Yuan et al. (2003).

2.2.2. Element analysis

Half of each aerosol sample and blank filters were digested at 170 °C for 4 hours in a high pressure Teflon digestion vessel with 3ml concentrated \(\text{HNO}_3\), 1ml concentrated \(\text{HClO}_4\), and 1 ml concentrated HF. The solutions were dried, and then diluted to 10 ml with deionized water (18 MΩ cm\(^{-1}\)). Fifteen elements (Al, As, Ca, Cd, Cu, Fe, Mg, Mn, Na, Ni, Pb, Sr, Ti, V, and Zn) were measured by an inductively coupled plasma optical emission spectroscopy (ICP-OES; SPECTRO, Germany). The detailed analytical procedures were described by Sun et al. (2004a) and Zhuang et al. (2001).

2.3. Nation-wide daily PM\(_{10}\) data

Air pollution index (API) data in 86 major cities (locations shown in Fig. 1a) over China were obtained from the data center of Ministry of Environmental Protection of China (http://datacenter.mep.gov.cn/). API was converted to PM\(_{10}\) concentration according to the following formula:

\[
C = C_{\text{low}} + [(I - I_{\text{low}})/(I_{\text{high}} - I_{\text{low}})] \times (C_{\text{high}} - C_{\text{low}}),
\]

Where \(C\) is the concentration of PM\(_{10}\) and \(I\) is the API value of PM\(_{10}\). \(I_{\text{high}}\) and \(I_{\text{low}}\), the two values that the most approaching to value \(I\) in the API grading limited value table, stand for the value larger and lower than \(I\), respectively; \(C_{\text{high}}\) and \(C_{\text{low}}\) represent the PM\(_{10}\) concentration corresponding to \(I_{\text{high}}\) and \(I_{\text{low}}\), respectively. It should be noted that API was recorded with a maximum value of 500, which corresponded to the PM\(_{10}\)
concentration of 600 µgm^{-3}.

2.4. Backward trajectory analysis

48 hours back trajectories of the air masses at both 250 m and 1000 m AGL (Above Ground Level) during dust days starting at Shanghai were computed by the HYSPLIT model (http://ready.arl.noaa.gov/hypub-bin/trajtype.pl?runtype=archive), using the meteorological data of GDAS (1 degree, global, 2006-present). Four trajectories ending at 00, 06, 12, and 18 LST were calculated for each day.

2.5. Model simulation

The WRF/CMAQ modeling system was applied to simulate the dust chemistry in this study. The Weather Research and Forecasting model (WRFv3.4) was used to produce the meteorology fields by digesting the reanalysis data from National Centers for Environmental Prediction (NCEP). The Community Multiscale Air Quality Modeling System (CMAQv5.0.1) was configured with the 2005 carbon bond gas-phase mechanism (CB05) and aerosol module AE6. The default CMAQ model doesn’t include dust chemistry reactions but only the dust emission module. We have implemented dust chemistry in the CMAQ model and corrected the dust emission module in our earlier work (Dong et al., 2015), which was used to simulate the heterogeneous reactions on dust during the long-range transport in this study. Briefly, the major developments included: (1) The default dust emission module in CMAQ was found to strongly underestimate the dust emissions. By removing the double counting of soil moisture in the default dust emission module and conducting a reanalysis of field data, the threshold friction velocities over various land covers were re-adjusted. (2) The source-dependent speciation profiles of dust particles from the Taklimakan and Gobi Desert were implemented based on field measurement data. (3) Thirteen dust heterogeneous reactions were implemented, including dust reactions with O_3, OH, H_2O_2, CH_3COOH, CH_3OH, CH_2O, HNO_3, N_2O_5, NO_2, NO_3, HO_2, and
SO2. The uptake coefficients of gases onto the surface of dust particles were taken from previous published studies in deserts of China. More details of the technical development of this dust emission and chemistry module in CMAQ and the evaluation of model performance can be found in Dong et al. (2015).

The modeling domain includes whole China with a horizontal grid resolution of 36 km×36 km and 34 vertical layers with a model top at 50 hPa. The chemical initial and boundary conditions for CMAQ were downscaled from the GEOS-Chem global model. Emissions inputs included anthropogenic emissions from Zhao et al. (2013) over China, biogenic emissions from MEGAN2.1 (Guenther et al., 2006), and biomass burning emissions from FLAMBE (Reid et al., 2009).

3. Results and Discussion
3.1. Spatiotemporal evolution of the 2010 spring dust storm over China

On March 19, 2010, a super dust storm outbroke over the Gobi Desert of Inner Mongolia, China and southern Mongolia. It was driven by a strong cold front and revealed evidently by the OMI (Ozone Monitoring Instrument) aerosol index (AI) from space detection (Tatarov et al., 2012). Along with the cold front, the dust plumes moved southeastward, invading extensive areas from Northern China to Southern China. Fig. 2 shows the daily PM10 concentrations over 86 cities of China from March 19 to 23, 2010, displaying the spatiotemporal evolution of this dust storm event. On March 20, daily PM10 concentrations of higher than 200 µgm⁻³ were observed over widespread areas of Northern China, close to the dust source regions. The PM10 concentrations of those heavily polluted cities, including Yinchuan (YC), Datong (DT), Hohhot (HHT), and Beijing (BJ) (red circles in Fig.1a,) reached the threshold of 600 µgm⁻³, the maximum value recorded by API (Fig. 2a). As the dust plumes transported, high levels of PM10 started to emerge over Central and Eastern China on March 21, when heavy pollution with daily PM10 concentrations higher than 420µgm⁻³ were observed over the Yangtze River Delta region as well as in Jiangxi,
Hunan, and Hubei provinces. Specifically, the PM$_{10}$ concentrations in Shanghai, Nanjing (NJ), Hefei (HF), Wuhan (WH), Hangzhou (HZ), and so on (blue circles in Fig. 1a) all reached the threshold of 600 µgm$^{-3}$ (Fig. 2c). On March 22 and 23, the dust plumes drifted towards Fujian and Guangdong provinces in Southern China, where PM$_{10}$ concentrations of over 600µgm$^{-3}$ were even observed in coastal cities such as Fuzhou (FZ), Xiamen (XM), and Shantou (ST) (pink circles in Fig.1a). The severe air pollution over China caused by this super dust storm lasted for ~4 days until March 23, resulting in significant impacts on the regional air quality and possible perturbation on regional meteorology.

3.2. Identification of two dust plumes with distinct transport pathways

Fig. 3a shows the time-height cross-section of aerosol depolarization ratios (DR) measured at the wavelength of 532 nm from March 19 to 23 in Shanghai. DR of dust particles is higher than pollution particles due to the non-sphericity of dust particles. Thus, aerosol DR is a useful parameter to identify dust events and a threshold value of 10% is usually used to distinguish dust from other types of particles (Shimizu et al., 2004). As shown in Fig. 3a, there were evidently two discontinuous periods with aerosol DR higher than 10%, consistent with the two peaks of PM$_{10}$ concentrations measured near the ground (Fig. 4c). The first dust episode (DS1) started from ~16:00 LST, March 20 to ~10:00LST, March 21 and the second dust episode (DS2) started from ~6:00 LST, March 22 to ~0:00 LST, March 23.

The 48-hours backward trajectories of the air masses during March 20-23 in Shanghai are shown in Fig. 2. It is interesting to note that the transport pathways of the dust plumes in DS1 and DS2 were distinctly different. In DS1, the dust plumes at both low altitudes (i.e. 250m denoted by the black lines in Fig. 2c-d) and high altitudes (i.e. 1000m denoted by the red lines in Fig. 2c-d) were mostly transported from the dust source regions in the Gobi Desert. This is one of the typical inland transport pathways of Asian dust, which passed over Northern China that is
characterized of intense anthropogenic emission rates, e.g. from Shanxi, Hebei, and Shandong provinces.

In DS2, the air masses reaching Shanghai are shown in Fig. 2e–h. From the starting of DS2 to the midday of March 22 (Fig. 2e-f), the backward trajectories were still mainly from the north but we noticed that the low altitude trajectories which originated from the Gobi Desert travelled over the Yellow Sea and East China Sea and then circled back to Shanghai. After the midday of March 22 (Fig. 2g-h), the trajectories became much shorter and restricted within the coastal and offshore areas. The low-altitude trajectories were mainly from the ocean and the high-altitude trajectories shifted from the south. As indicated by the surface observation (Fig. 4a), the southeasterly winds prevailed until ~18:00 LST, March 22 when PM$_{10}$ climbed to reach its peak value in DS2, quite different from DS1 when the northerly winds dominated. The CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) transect at around 13:00 – 13:30 LST, March 21 specified that the major aerosol type was dust over the East China Sea and South China Sea (Fig. S1). Therefore, it was evident that DS2 passed over the coastal regions of Eastern China and the ocean and it should be a humid dust plume.

As visualized in Fig. 3, the two dust plumes were mostly transported at low altitudes below 1km. It is expected that this type of transport would benefit for the mixing and interactions between dust particles and anthropogenic pollutants, as the anthropogenic emissions were mainly trapped within the boundary layer. Transport pathway is one of the most important factors accounting for the evolution of dust particles and the resulting environmental effects (Zaizen et al., 2014; Zhang et al., 2010). In this super dust storm, the two discontinuous dust plumes reaching Shanghai were distinctly different in their transport pathways, providing a great opportunity to study the chemical evolution of dust particles under different transport conditions.

3.3. Distinct behaviors of gaseous pollutants between two dust episodes
Fig. 4 illustrates the different behaviors of the two identified dust episodes by plotting the temporal variations of emission precursors (i.e. SO₂ and NO₂) as well as PM₁₀ and crucial meteorological parameters. As shown in Fig. 4a, the winds prevailed from the south before the onset of DS1 at ~10:00 LST, March 20. During this period, SO₂ and NO₂ concentrations were relatively high with mean concentrations of 39 ± 19 and 70 ± 25 µg m⁻³, respectively. In the meantime, PM₁₀ was at a moderate level of 112 ± 54 µg m⁻³, suggesting the dominance by anthropogenic emissions. It was observed that the peak hourly concentrations of SO₂ and NO₂ up to 97 µg m⁻³ and 116 µg m⁻³ occurred just a few hours before the sharp increase of PM₁₀. This was due to that air pollutants north of Shanghai were pushed by the cold front and accumulated before the dust plumes invaded (Guo et al., 2004), which was also evident from the time difference between aerosol backscattering coefficients and depolarization ratios as shown in Fig. 3.

From 10:00 LST, March 20 to 11:00 LST, March 21, the winds shifted from the north and northwest accompanied with a quick drop of relative humidity (RH) with a minimum of 25% at 21:00 LST, March 20, indicating the invasion of a strong cold front from Northern China. Correspondingly, PM₁₀ abruptly climbed since 16:00 LST and reached 1000 µg m⁻³ (a maximum value of 1000 µg m⁻³ was set in the TEOM 1405D monitor) within four hours. This high level of PM₁₀ lasted for ~11 hours till 04:00 LST, March 21. During this dust episode, the concentrations of SO₂ and NO₂ decreased substantially and the lowest values of 17 µg m⁻³ and 27 µg m⁻³ (Fig. 4c) were recorded at 04:00 LST, March 21, due to the strong dilution effect of dust plumes. The ratios of SO₂/PM₁₀ and NO₂/PM₁₀ were as low as 0.04 ± 0.02 and 0.06 ± 0.03 in DS1 compared to that of 0.30 ± 0.05 and 0.57 ± 0.28 before DS1. After 12:00 LST, March 21, the wind direction started to shift again from the southeast with an increase of RH. The PM₁₀ concentrations quickly decreased below 200 µg m⁻³ within five hours, indicating the pass of the first dust episode over Shanghai.

The second wave of dust (DS2) commenced after about half a day as indicated by
the temporal variation of PM$_{10}$ climbing quickly since 06:00 LST, March 22. PM$_{10}$ reached its peak of 530 µgm$^{-3}$ observed at ~18:00LST, March 22, much lower than the peak values during DS1. It should be noted that during the growth of DS2 (i.e. from 06:00 to 18:00 LST), southerly winds sustained until 18:00 LST, March 22, opposite to DS1. As discussed above, DS2 had gone through the maritime environment during most of its transport trajectory. This can be corroborated by the moderately high RH (Fig. 4b). After 18:00 LST, March 22, PM$_{10}$ underwent a quick decline followed by the prevailing northeast winds and continuously elevated RH. This suggested the sea breezes after 18:00 LST were almost free of dust particles and acted as cleaner for the dust pollution that was previously accumulated. Compared to the mean RH of 46% ± 18% in DS1, RH in DS2 was much higher of 69% ± 8%. This was mainly attributed to the different transport pathways of the two dust episodes.

One interesting phenomenon that has been rarely observed was that the temporal variations of SO$_2$ and NO$_2$ varied fairly consistent with that of PM$_{10}$ in DS2 (Fig. 4c), quite different from DS1 and previous studies (Fu et al., 2010; Guo et al., 2004) that dust usually had a clean effect on the local gaseous pollutants, causing inverse relationship between SO$_2$/NO$_2$ and PM$_{10}$. In this case, NO$_2$ reached its maximum hourly concentration of 131µgm$^{-3}$ along with the maximum PM$_{10}$ in DS2, the highest during the whole study period. This probably indicated that the dust particles in DS2 were externally mixed or “coated” with abundant gaseous pollutants. As a result, the SO$_2$/PM$_{10}$ and NO$_2$/PM$_{10}$ ratios reached 0.11 ± 0.03 and 0.20 ± 0.04 in DS2, respectively, much higher than those in DS1 and in a super dust day of April 2, 2007 in Shanghai (Fu et al., 2010), revealing that the dust plumes in DS2 were much more polluted.

In the following sections, we will investigate deeply into the chemical characteristics and evolution of the two dust episodes.

3.4. Chemical evolution of dust aerosols
3.4.1. Pollution elements significantly enhanced in dust periods

Table 1 lists the concentrations of measured elements in TSP before, during, and after the two dust episodes in Shanghai. D and N represent the samples that collected in daytime (~8:00 to ~20:00 LST) and nighttime (~20:00 to 8:00 LST in the next day), respectively. NDS represents the non-dust days from March 25 to 27 and the mean concentrations are shown in Table 1. To identify whether these elements were mainly derived from the crustal source or anthropogenic sources, enrichment factors of elements were calculated. Enrichment factor (EF) is defined as

$$EF = \frac{(X/X_{Ref})_{aerosol}}{(X/X_{Ref})_{crust}}$$

where \((X/X_{Ref})_{aerosol}\) is the mass ratio of a given element X to the reference element in aerosol and \((X/X_{Ref})_{crust}\) is the ratio in the crust (Lida, 2006), and Al is the reference element used in this study. EFs of Al, Cu, Ca, Fe, Mg, Na, Mn, Ti, Sr, Ni, and V were calculated to be lower than 5, indicating these eleven elements were mostly from the crustal source, while EFs of As, Cd, Pb, and Zn were higher than 10 even during the dust periods (Fig.5c), indicating these four elements were significantly influenced by anthropogenic sources. Hence, Al, Cu, Ca, Fe, Mg, Na, Mn, Ti, Sr, Ni, and V were classified as “crustal elements” while As, Cd, Pb, and Zn were classified as “pollution elements”. Due to the dilution effect of dust plumes, EFs of the four pollution elements decreased from ~100-150 in NDS to ~10-50 in DS1 and ~20-115 in DS2.

As shown in Table 1, the concentrations of the crustal elements before the dust event (19N) were as low as those in NDS, but substantially increased in DS1 and DS2, about 3-13 and 1-7 folds of those in NDS, respectively. The highest concentration of Al, widely used as a tracer for dust, reached 67.5 µgm\(^{-3}\) in DS1 (20N), comparable to the Al concentration in the super dust day of April 2, 2007 in Shanghai (Fu et al., 2010). The highest concentrations of crustal elements in DS1 (20N) were ~2 folds of those in DS2 (22D), corroborating that the intensity of DS1 was stronger than in DS2.

As for the pollution elements, the non-crustal part (nc-) of As, Cd, Pb, and Zn were estimated as nc-X = X – Al × (X/Al)\(_{crust}\). The concentrations of nc-As, nc-Cd, nc-Pb,
and nc-Zn increased in both DS1 and DS2, about 1-4 folds of those in NDS (Fig. 5a). As these elements were mainly derived from anthropogenic sources such as coal combustion, industrial processing, vehicle emissions, etc., their enhancement indicated that abundant pollutants had been transported to the downwind regions along with the dust plumes. The mean concentrations of nc-As, nc-Cd, nc-Pb, and nc-Zn were 23.5, 3.6, 154.2, and 580.1 ng m\(^{-3}\) in DS2, higher than those of 18.3, 3.1, 119.7, and 447.4 ng m\(^{-3}\) in DS1, indicating DS2 was more polluted. Consistently, the mass ratios of nc-As, nc-Cd, nc-Pb, and nc-Zn in TSP were 5.1 \(\times\) 10\(^{-3}\)%, 7.2 \(\times\) 10\(^{-4}\)%, 3.1 \(\times\) 10\(^{-2}\)%, and 1.2 \(\times\) 10\(^{-3}\)% in DS2, higher than those of 1.9 \(\times\) 10\(^{-3}\)%, 3.7 \(\times\) 10\(^{-4}\)%, 1.5 \(\times\) 10\(^{-2}\)%, and 6.2 \(\times\) 10\(^{-3}\)% in DS1 (Fig. 5b).

3.4.2. Crustal vs. secondary water-soluble ions

Fig. 6 shows the evolution of major water-soluble ions in TSP during the whole dust event. The concentrations of Na\(^{+}\), K\(^{+}\), Mg\(^{2+}\), and Ca\(^{2+}\) increased the most among all the ions in both DS1 and DS2 due to their crustal origin, ~2-5 and ~2-3 folds of those in NDS. However, the mass ratios of these ions in TSP generally decreased in the dust periods (Fig. 6d), as the masses of dust particles were dominated by water-insoluble matters. Different from NDS, the concentrations of Ca\(^{2+}\) exceeded NH\(_4\)\(^{+}\) and ranked as the most abundant cation in the dust periods. This was because Asian dust particles are rich of calcium carbonate (Wang et al., 2005), which could react with acidic nitrogen and sulfur compounds to form Ca(NO\(_3\))\(_2\) and CaSO\(_4\). It has been found Ca-rich particles in Asian dust transported to Japan were mostly in the spherical shape (Zaizen et al., 2014), and many of the spherical Ca-rich particles contained sulfur and nitrogen compounds (Matsuki et al., 2006; Zaizen et al., 2014) due to the heterogeneous reactions. It is noted that the concentrations of Ca\(^{2+}\) in DS2 were comparable to that in DS1 (Fig. 6b), although the intensity of DS1 was much stronger than DS2. In addition, the mass ratios of Ca\(^{2+}\) in TSP were even higher in DS2 than in DS1 (Fig. 6d). Collectively, these results indicated that more calcium in
its soluble form was produced via the reactions between calcium carbonate and acids. This could be also revealed by the ratio of Ca\(^{2+}\)/Ca that was higher in DS2 (0.2-0.5) than in DS1 (0.1-0.2) (Fig.7a). In DS2, the dust plumes travelled over the ocean and carried higher amount of water vapor as well as SO\(_2\) and NO\(_2\) than DS1 as discussed earlier. In this regard, the heterogeneous reaction between calcium carbonate and acidic gases was enhanced more in DS2, resulting in a higher fraction of calcium carbonate from dust particles that could be transformed to soluble calcium.

Cl\(^-\) during the dust periods was moderately enhanced by ~1-2 folds compared to NDS, indicating that Cl\(^-\) was also impacted by the invasion of dust plumes. As shown in Fig. 6a & 6c, both the concentrations of Cl\(^-\) and its mass ratios in TSP were higher in DS2 than in DS1. As the air masses of DS2 passed over the ocean, part of the high Cl\(^-\) concentrations should be attributed to the contribution from sea salts. The average equivalent ratio of Cl\(^-\) to Na\(^+\) ([Cl\(^-\)]/[Na\(^+\)]) was 1.65 in NDS and 2.17 in 19N, higher than the ratio in seawater (1.17), indicating that anthropogenic sources such as coal combustion (Sun et al., 2014; Yao et al., 2002) contributed significantly to Cl\(^-\) in the non-dust days. The [Cl\(^-\)]/[Na\(^+\)] ratio dropped to ~1.0 in DS1 as massive continental aerosols invaded Shanghai, while in DS2 the ratio was elevated to 1.5. Thus, both sea salts and anthropogenic sources should contribute to the particulate Cl\(^-\) in DS2.

As for the secondary inorganic ions (i.e. SO\(_4^{2-}\), NO\(_3^-\), and NH\(_4^+\)), their concentrations in TSP were elevated to be 16.2, 15.4, and 7.2\(\mu\)gm\(^-3\) in the daytime of March 20 (20D) in DS1. The sampling time of 20D was from ~8:00 to 20:00 LST, including the short period of pollutants accumulation before the arrival of DS1. As a result, the high concentrations of SO\(_4^{2-}\) and NO\(_3^-\) measured in 20D were contributed from both local and transported sulfate and nitrate, as well as from the heterogeneous reactions on dust particles. In the nighttime of March 20 (20N), the concentrations of NO\(_3^-\) and NH\(_4^+\) sharply decreased by 57.8% and 43.2%, respectively. Although SO\(_4^{2-}\) also decreased, its reduction extent was much weaker of 27.8% and was still ~2 folds of that in NDS (Fig. 6a). Nitrate formation on dust is strongly dependent on the
ambient conditions. Low temperature, low relative humidity, strong wind and low concentrations of pollution gases did not favor the heterogeneous reaction (Huang et al., 2010a; Yuan et al., 2008). Hence, strong dust events usually diluted the concentrations of nitrate (Duvall et al., 2008; Huang et al., 2010a; Wang et al., 2006). As for the sulfate formation on dust, SO$_2$ could interact with various mineral components of dust particles to produce sulfate, and sulfate became mixed with the dust first competing with nitrate (Dupart et al., 2012; Sullivan et al., 2007). Hence, although the secondary aerosol components were diluted by the invasion of dust, the formation of sulfate could still be compensated by the heterogeneous reaction on dust to some extent.

In the daytime of March 22 (22D) during DS2, the concentrations of SO$_4^{2-}$ and NO$_3^-$ reached the highest during the study period with values of 16.6 and 19.3 µgm$^{-3}$, respectively. Although SO$_4^{2-}$ and NO$_3^-$ decreased in the nighttime of March 22 (22N), their levels were still higher than the non-dust days (Fig. 6a). Also, the mass ratios of SO$_4^{2-}$ and NO$_3^-$ in TSP were higher in DS2 than DS1 (Fig. 6c). These results indicated that SO$_4^{2-}$ and NO$_3^-$ were more favorably formed in DS2. It was observed that the concentration of NO$_3^-$ was even higher than that of SO$_4^{2-}$ in both daytime and nighttime samples during DS2, showing a contrary behavior to DS1. As discussed above, the formation of nitrate on dust strongly depends on the ambient conditions.

During DS2, RH was at a moderately high level, enhancing the efficiency of aqueous processing on the particles. In addition, the abnormally high concentrations of NO$_2$ (Fig. 4c) suggested the emission precursors were sufficient for the production of nitrate. Finally, high concentrations of nitrate during DS2 may be partially attributed to the reaction between sea salts and nitric acid (Hsu et al., 2014; Huang et al., 2010a) as the dust plumes travelled over the ocean. The formation of secondary aerosol during DS2 should have involved the complex interactions between pollutant precursors, dust, and sea salts.

Fig. 7b shows the equivalent ratio of the total anions to the total cations (A/C) in
TSP. The A/C ratio dropped to ~0.5 in DS1 from ~0.9 in 19N, which was attributed to the existence of abundant CO$_3^{2-}$ in dust particles that can’t be detected by Ion Chromatography (Huang et al., 2010a). The A/C ratio reached 0.85 and 0.80 in 22D and 22N in DS2, respectively, much higher than those in DS1, suggesting that more fraction of CaCO$_3$ in the dust particles in DS2 was transformed to CaSO$_4$ and Ca(NO$_3$)$_2$ than in DS1, consistent with the higher Ca$^{2+}$/Ca ratio in DS2 than in DS1 (Fig. 7a). The equivalent ratio of NH$_4^+$ to the sum of SO$_4^{2-}$ and NO$_3^-$ ([NH$_4^+$]/[SO$_4^{2-}$+NO$_3^-$]) in TSP also indicated that the heterogeneous reactions between carbonate and acid gases were more promoted in DS2 compared to DS1. As shown in Fig. 7c, the [NH$_4^+$]/[SO$_4^{2-}$+NO$_3^-$] ratio was slightly higher than one in the non-dust days, indicating that SO$_4^{2-}$ and NO$_3^-$ could be completely neutralized by NH$_4^+$. In DS1, the [NH$_4^+$]/[SO$_4^{2-}$+NO$_3^-$] ratio was 0.68 and 0.65 in 20D and 20N, while in DS2 it was even lower of 0.45 and 0.55 in 22D and 22N, respectively. Apparently, NH$_4^+$ was insufficient for completely neutralizing SO$_4^{2-}$ and NO$_3^-$ in both dust episodes, particularly in DS2. The ammonium deficiency was also observed in Kinmen and Zhuhai (Hsu et al., 2014), an island site and a coastal site, respectively, during this super dust storm. We further investigated the [NH$_4^+$+Ca$^{2+}$]/[SO$_4^{2-}$+NO$_3^-$] ratio. As shown in Fig. 7d, with the addition of Ca$^{2+}$, sulfate and nitrate had been completely neutralized, implying the important role of alkaline calcium as the medium of dust heterogeneous reactions. By estimating the neutralization efficiency of Ca$^{2+}$ (NE$_{Ca}$) as NE$_{Ca}$ = 1 - [NH$_4^+$]/[SO$_4^{2-}$+NO$_3^-$], the average value of NE$_{Ca}$ in DS1 and DS2 was 0.34 and 0.50, respectively. The higher NE$_{Ca}$ in DS2 also suggested the chemical processing via dust was efficient under the environmental conditions such as DS2 in this study.

3.5. Vertical evolution of dust particles

In both DS1 and DS2, dust aerosols were mostly refrained below the altitude of 1km. Strong vertical gradients of DR and BSC were observed (Fig. 3). To
quantitatively investigate the evolution of vertical dust profiles, we selected four

typical episodes as follows: 1. 9:00-15:00 LST of March 20: an episode right before

the arrival of DS1; 2. 16:00 - 17:45 LST of March 20: an episode before the onset of

maximum hourly PM$_{10}$ concentrations in DS1; 3. 18:00 LST of March 20 - 04:45 LST

of March 21, an episode that covers the highest PM$_{10}$ concentrations hours

(>1000µg/m$^3$) in DS1; 4. 6:00 - 18:00 LST of March 22: an episode in DS2.

As shown in Fig. 8a, the mean BSC between 9:00 - 15:00LST, March 20 ranged

from around 0.005 to 0.02 sr$^{-1}$km$^{-1}$ from near ground level to 1km. During this period,
the mean DR was relatively low of ~ 0.02 - 0.08, suggesting spheric particles, i.e.
pollution particles dominated. Accordingly, the contribution of dust to the total
extinction was less than 20% (Fig. 8c). As discussed earlier, the high values of light
extinction caused by pollution aerosol before the onset of DS1 were due to the
pre-accumulation of local and transported pollutants brought by the invaded cold front.

On the pathway of DS1 (Fig. 2c), the air masses travelled over large areas with
intense anthropogenic emissions and thus a large quantity of pollutants could be
pushed to the downstream areas and accumulated before the dust plume arrived.

As DS1 invaded, BSC further increased and reached 0.025 sr$^{-1}$km$^{-1}$ near the surface
level (Fig. 8b-c). In the meantime, the values of DR were elevated to be higher than
10% from near the ground to ~ 1km (Fig.8f-g). When PM$_{10}$ reached its highest
concentrations (>1000µg/m$^3$), the mean DR was ~20-25% extending from near the
ground to ~700m (Fig.8g). However, compared to the DR value of ~30–35% for the
relatively pure Asian dust (Murayama et al., 2003; Sakai et al., 2003), DR of DS1 was
still lower, suggesting the mixing of dust with pollution at a certain extent. As shown
in Fig. 8b-c, the extinction coefficient of dust particles obviously overwhelmed that of
pollution particles and could be up to 0.82 km$^{-1}$ near the surface level, while that of
pollution particles dropped to less than 0.1 km$^{-1}$. From near the ground to ~700m, no
significant vertical gradients of DR and dust ratio (the ratio of the dust extinction in
the total extinction of aerosols) were observed. This indicated DS1 had a “top-down”
effect on modifying the bulk aerosol optical properties in the lower troposphere in this case. Within this altitude range, dust could account for ~80 - 90% of the total aerosol extinction coefficients while only a minor fraction of ~10 - 20% was attributed to the pollution aerosols. Above 700m, DR and the dust ratio quickly deceased, suggesting the impact of dust on the light extinction was much weakened.

In DS2, the maximum aerosol BSC and DR averaged between 6:00 and 18:00 LST, March 22 was ~0.015 sr$^{-1}$km$^{-1}$ and 18%, respectively, observed at an altitude of ~300m. Different from DS1, both DR and the dust ratio had a significant vertical gradient in DS2. As showed in Fig. 8h, DR decreased quickly from its maximum value at ~250m to 5% at ~750m. Correspondingly, the dust ratios decreased from 88% to 25%. Moreover, the vertical profiles of dust and pollution extinction coefficients showed distinctly different behaviors during this period. As shown in Fig. 8d, the maximum extinction coefficient of dust particles (~0.55 km$^{-1}$) showed at the altitude of ~250m, while that of pollution particles (~0.25 km$^{-1}$) appeared at a higher altitude of ~800m. As compared to DS1 (Fig. 8b-c), aerosol extinction caused by spheric particles in DS2 showed similar magnitudes below ~270m. Above this altitude, aerosol extinction caused by spheric particles in DS2 gradually increased while that in DS1 varied relatively stable with the altitudes. The enhancement of pollution particles in the middle layer during DS2 should be mainly attributed to its unique transport pathway. As shown in Fig. 2e, substantial air masses in the upper layer transported from the south and may bring more moisture. The sounding data at a meteorology station (31.40°N, 121.46°E) in Shanghai supported this statement as shown in Fig. S2.

Opposite to the low relative humidity (RH) and its decreasing trend with altitude in DS1, RH in DS2 (measured at 8:00 and 20:00 LST of March 22, respectively) showed much higher values and an increasing trend with altitude (Fig. S2). This phenomenon corroborated our discussions above that the meteorological conditions were more favorable for promoting the dust chemistry in DS2. As a result, the secondary aerosol
formation via heterogeneous reaction yielded stronger aerosol extinction in DS2, especially in the middle and upper layers through 270m till the top (2km) where the aerosol extinctions caused by spheric particles were still significant. On the other hand, due to the higher humidity in DS2, the soluble aerosol components should undergo stronger hygroscopic growth and thus partly explain the structure of vertical profile of spheric particles as shown in Fig. 8d. It is commonly regarded that in a dry and less oxidative environment that dust storms are usually associated with, the formation and growth of secondary aerosols are often depressed. However, under certain favorable conditions, new particle formation during dust events could be still discernible (Nie et al., 2014). In this study, vertical profiles of crucial meteorological parameters, pollutant precursors and particle numbers were not available for diagnosing the new particle formation. Tethered balloon-based measurement (Li et al., 2015) could be a good platform for investigating the particle formation during dust events at different altitudes in the future research.

3.6 Impact of dust chemistry on regional air quality

Chemical transport modeling (Methods in Section 2.5) was utilized to assess the impact of dust chemistry on the perturbation of air quality at the regional scale. The model performance of CMAQ with improved dust module has been evaluated against various observational datasets and it was demonstrated that the model has relatively good capability in capturing both magnitudes and temporal variability of bulk aerosol (e.g. PM$_{10}$, AOD) during the spring season over China (Dong et al., 2016). It has to be noted that the model only simulated the aerosol size up to 10 µm while the observation of aerosol chemistry included all the sizes (i.e. TSP), hence the mismatch of size distribution between the model and observation precluded the evaluation of the simulated aerosol chemical species in this study. In the following discussions, we focused on the qualitative assessment of the impact of dust chemistry on the regional air quality.
Fig. 9 shows the spatial distribution of simulated mineral aerosols from March 20 – 22, 2010, respectively. Accordingly, we show the spatial distribution of Ultraviolet Aerosol Index (UVAI) retrieved from OMI during the same period. UVAI is sensitive to absorbing aerosols, i.e. black carbon and mineral dust (Torres et al., 2007). Hence, the comparison between simulated mineral aerosols and observed UVAI could illustrate how the transport of dust was reproduced. As seen from Fig. 9, high values of UVAI were always observed over the Indo-China Peninsula and this was ascribed to the black carbon aerosols emitted from strong biomass burning over this region during the spring season (Fu et al., 2012; Huang et al., 2013; Tsay et al., 2013). What we concern are the dust and its downwind regions over East Asia. On March 20, high UVAI values stretched from the Gobi Desert to the North China Plain and the Yangtze River Delta. This transport pathway was relatively well reproduced by the model as we can see high concentrations of mineral aerosols over similar areas as well. On the following day of March 21, the satellite observation illustrated the movement of the high UVAI zones further southward and the drifting of dust plumes off the coastline of East China (Fig. 9c). Accordingly, the simulation showed similar behavior with strengthened concentrations of mineral aerosols over the Gobi Desert and downwind areas of the Yangtze River Delta (Fig. 9d). On March 22, although UVAI signals were absent over most areas of East China due to the satellite swath, high UVAI values could still be observed over the tip of the Yangtze River Delta and the East China Sea. This is also relatively consistent with the model simulation that high concentrations of mineral aerosols hovered over the coastlines from YRD to the Taiwan Strait. Overall, we demonstrated that the model is capable of capturing the spatial distribution of dust during the long-range transport.

Since we specifically focus on the interaction between dust and anthropogenic pollutants in this study, we have performed two simulations, i.e. one with dust emissions but without dust chemistry and the other one with dust chemistry. The impact of dust chemistry on aerosol chemical components could thus be quantified via
the difference between these two simulations. Fig. 10a&c shows the spatial distribution of sulfate via the formation pathway of dust heterogeneous reactions, as well as for nitrate in Fig. 10b&d. Different from the spatial distribution of mineral aerosols (Fig. 9a-c), the formation of sulfate and nitrate via dust chemistry mainly occurred over Northeastern China. This is expected that although the major source region of mineral dust is from the Gobi Desert in Northern China, less anthropogenic emission sources existed there, hence relatively weak atmospheric chemical processing was simulated over the dust source region. On the other hand, drier climate in Northern China also suppressed the extent of heterogeneous reactions on the surface of dust. Hence, the strongest impact from dust heterogeneous reactions on the formation of secondary aerosols occurred in those populous areas where the interaction of anthropogenic precursors and dust was the strongest.

As for sulfate, the simulated concentration over Shanghai averaged 8.1, 3.1, and 8.5 μgm⁻³ from March 20 – 22, respectively. This temporal variation corresponded to that from observation as discussed in Section 3.4.2. While it has to be noted again that the simulated aerosol species contained particulate sizes less than 10 μm, close match between the simulation and observation was not expected. As we compare DS1 (Fig. 10a) and DS2 (Fig. 10c), the simulated sulfate during DS2 was evidently more intense than that during DS1 at a larger geographic region, which was fairly consistent with the observation. Since the daily emission rates digested by the model were almost constant during this period, meteorological conditions should be the determining factor, of which elevated humidity during DS2 has been diagnosed as the most important factor responsible for stronger dust chemistry.

As for nitrate, its spatial distribution pattern was as similar as that of sulfate at a certain extent. The simulated concentration of nitrate averaged 3.1, 2.3, and 5.2 μgm⁻³ over Shanghai from March 20 – 22, respectively. Although the simulation results and observational data couldn’t be statistically inter-compared due to the size difference as stated above, we think the simulated nitrate should be largely underestimated. The
ratio of simulated nitrate between DS2 and DS1 was 1.7, similar to that of ~1.6 from observation. This suggested the favorable meteorological conditions (e.g. higher humidity) on facilitating the dust heterogeneous reaction during DS2 have been accounted by the model. In this regard, we ascribe the low-biased nitrate mainly to several aspects. First, the NOx emissions could have been underestimated. As indicated in Dong et al. (2015) which used the same anthropogenic emission inventory as this study, the model showed some underestimation of the total NO2 columns as compared to the OMI observation, especially over Northeastern China (See Fig. 8 in Dong et al.(2015)). Secondly, HNO3, N2O5, NO2 and NO3 were the major precursors of nitrate as implemented in the dust module. Of which, reactions via HNO3 and N2O5 were the dominant pathways of nitrate formation due to their relatively high uptake coefficients on the dust. It was possible that the formation of HNO3 was underestimated, thus lowering the production of nitrate. However, this is just a guess as observation of gaseous HNO3 was not available in this study. Lastly, we think the underestimation of nitrate may be due to the omission of nitrate processing on the surface of sea salt, which was especially important for costal cities such as Shanghai (Buseck and Posfai, 1999).

4. Conclusions

During March 19-23, 2010, a super dust storm swept extensive areas over China from Northern China to Southern China. Two separate dust periods were observed in Shanghai, one from ~16:00 LST, March 20 to ~10:00 LST March 21 (DS1) and the other from ~6:00 LST, March 22 to ~0:00 LST, March 23 (DS2). In DS1, the dust plumes mostly transported over those areas characterized of high pollution emissions in Northern China, while the dust plumes in DS2 transported over the coastal regions of Eastern China with higher RH. The ratios of SO2/PM10 and NO2/PM10 were up to $0.11 \pm 0.03$ and $0.20 \pm 0.04$ in DS2, much higher than the values in DS1. In addition, the concentrations of typical elemental species such as As, Cd, Pb, and Zn were also
more enriched in DS2 than in DS1, indicating the stronger mixing extent between dust and primary anthropogenic emissions in DS2. Due to the higher level of gaseous pollutant precursors associated with moderate relative humidity, \( \text{SO}_4^{2-} \) and \( \text{NO}_3^- \) exhibited higher concentrations in DS2 than in DS1. The higher \( \text{Ca}^{2+}/\text{Ca} \) ratio and lower \( [\text{NH}_4^+]/[\text{SO}_4^{2-}+\text{NO}_3^-] \) ratio in DS2 suggested the heterogeneous reactions between calcites and acid gases were significantly promoted in DS2. Particularly, more \( \text{NO}_3^- \) than \( \text{SO}_4^{2-} \) was formed in DS2, probably due to the high concentrations of \( \text{NO}_2 \) and the complex reaction among dust, seasalts, and nitric acid. Vertical profiles of aerosol optical properties from Lidar measurement retrieved high depolarization ratios of 20-25% extending from near the ground to \(~700\)m in DS1 and it was estimated that dust dominantly accounted for \(~80-90\)% of the total aerosol extinction. In DS2, the vertical structure of aerosol changed dramatically from that of DS1. The maximum extinction coefficient of dust particles emerged at the altitude of \(~250\)m in DS2, while that of pollution particles showed at a higher altitude of \(~800\)m. The abnormal increase of humidity as a function of altitude from the southerly winds explained this phenomenon due to promoted heterogeneous reactions on dust particles and the subsequent aerosol hygroscopic growth.

Simulated mineral aerosols were compared to the remote sensing UV AI from satellite, showing consistent spatial patterns between model and observation. By applying the dust scheme with explicit chemistry mechanisms, evident impacts of dust heterogeneous reactions on secondary aerosol formation were reproduced over widespread areas of Northeastern China where the anthropogenic emissions were intensive. The sulfate formation was relatively well simulated while the nitrate formation on dust was believed to be largely underestimated. More research on laboratory kinetic studies of nitrate dust chemistry is suggested. Also, the feedback between dust chemistry and regional climate change needs to be investigated in the future.
Acknowledgment

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Table 1 Concentrations of the measured elements (μg/m³) in TSP before, during, and after the dust periods from March 19 to 22 and in non-dust days (NDS) in Shanghai. N, D, and NDS represent the nighttime samples collected from 20:00 to 8:00 in the next day, the daytime samples collected from 8:00 to 20:00, and the average of the non-dust samples collected from March 25 to 27, respectively.

<table>
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<tr>
<th></th>
<th>DS1</th>
<th>DS2</th>
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</thead>
<tbody>
<tr>
<td></td>
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<td>20 D</td>
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<tr>
<td>TSP</td>
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<tr>
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Fig.1 (a) Locations of the 86 major cities over China. The cities with PM$_{10}$ concentrations up to 600µg m$^{-3}$ on March 20, 21, and 22 were marked by red, blue, and pink circles, respectively. (b) The site location of the ground-based measurement in Shanghai.
Fig. 2. Daily PM$_{10}$ concentrations (mg m$^{-3}$) in 86 major cities over China and 48 hours backward trajectories of the air masses at both 250m (black lines) and 1000m (red lines) above ground level over Shanghai during March 19-23, 2010. Four trajectories ending at 00, 06, 12, and 18 LST are computed for each day.
Fig. 3 Time-height cross-section of (a) depolarization ratios and (b) attenuated aerosol backscattering coefficients measured by a Lidar at the wavelength of 532 nm during March 19-22, 2010.
Fig. 4 Time-series of PM$_{10}$, SO$_2$, NO$_2$, and meteorological factors including wind conditions, relative humidity (RH), and ambient temperature (T) in Shanghai during March 19-23, 2010.
Fig. 5 Variations of (a) the mass concentration ratio of each sample to the NDS sample for nc-As, nc-Cd, nc-Pb, and nc-Zn, (b) the mass ratios, and (c) enrichment factors of pollution elements As, Cd, Pb, and Zn in TSP during March 19-22 and non-dust days (NDS). D, N, and NDS represent the daytime samples collected from 8:00 to 20:00 LST, the nighttime samples collected from 20:00 to 8:00 LST in the next day, and the average of the non-dust samples collected from March 25 to 27, respectively.
Fig. 6 Variations of the concentrations of water-soluble ions and their mass ratios in TSP during March 19-22 and NDS, 2010.
Fig. 7 Variations of (a) the ratios of Ca$^{2+}$/Ca, (b) the equivalent concentrations of the total anions to the total cations (Anion/Cation), (c) the equivalent concentrations of NH$_4^+$ to the sum of SO$_4^{2-}$ and NO$_3^-$ ([NH$_4^+$]/[SO$_4^{2-}$+NO$_3^-$]), and (d) the equivalent concentrations of the sum of NH$_4^+$, Ca$^{2+}$, and Mg$^{2+}$ to the sum of SO$_4^{2-}$ and NO$_3^-$ ([NH$_4^+$ + Ca$^{2+}$ + Mg$^{2+}$]/[SO$_4^{2-}$+NO$_3^-$]) in TSP during March 19-22 and NDS, 2010.
Fig. 8 Vertical profiles of the average extinction coefficients of dust and pollution particles (km$^{-1}$), attenuated backscattering coefficient (BSC, sr$^{-1}$km$^{-1}$), depolarization ratios (DR, unitless), and the ratio of the dust extinction in the total extinction (Dust ratio, unitless) in four periods of 9:00 - 15:00 of March 20 (before DS1), 16:00 - 17:45 of March 20 (before the highest PM$_{10}$ concentration in DS1), 18:00 of March 20 - 04:45 of March 21 (during the highest PM$_{10}$ concentration in DS1), and 6:00 - 18:00 of March 22 in DS2.
Fig. 9. The spatial distribution of OMI Ultraviolet Aerosol Index (unitless) and simulated mineral aerosols (µgm⁻³) from March 20 – 22, 2010, respectively.
Fig. 10. Simulated $\text{SO}_4^{2-}$ and $\text{NO}_3^-$ ($\mu g/m^3$) from dust heterogeneous reactions during DS1 and DS2.